

## Supplemental Information

### Secondary organic aerosol coating formation and evaporation: Chamber studies using black carbon seed aerosol and the Single-Particle Soot Photometer

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### Measurement of angularly-resolved light scattering from single particles

The prototype instrument used in this study to measure particle sphericity was designed by MSP Corporation (Shoreview, MN, USA) to measure angularly-resolved light scattering from single particles. Air with particles enters the instrument and is guided towards the detection block by filtered sheath air and a focusing nozzle. The stream of particles is focused to align with the center of the laser beam in the detection block. The sheath and aerosol flows are feedback-controlled to maintain constant operating conditions throughout a sampling period. The laser is a 100 mW Diode Pumped Green CrystaLaser (Reno, NV, USA) at a peak wavelength of 532 nm. Lenses tightly focus the laser beam into the detection block, and a quarter-wave plate transforms the linearly polarized laser light into circular polarization inside the detection block. At its intersection with the particle stream, the Gaussian laser beam profile is approximately 440  $\mu\text{m}$  in diameter. The sphericity signal is detected with lenses placed on the end of 16 fiber optic cables attached to the detection block. Eight of the lenses are placed axisymmetrically about a polar angle of  $50^\circ$  in the forward scattering direction, and the other eight are placed axisymmetrically about a polar angle of  $130^\circ$ . The other ends of all the fiber optic cables are mounted in front of a Hamamatsu H8711 4x4 multianode photomultiplier tube (PMT) assembly. Each of the fiber optic cables is focused toward one of the 16 channels in this PMT assembly. In this study, only the eight signals from a polar angle of  $50^\circ$  are used to determine particle sphericity.

Particle sphericity is determined by quantifying the degree to which the light scattered onto the eight detectors at a fixed polar angle is uniform. Owing to artifacts introduced by the different optical fiber characteristics on each of these channels, the signal must be adjusted on each channel independently of the other channels. This adjustment is done by normalizing the signal,  $\Psi$ , on each detector by the median response of that detector for a fixed amount of time before and after each

individual measurement by

$$\hat{\Psi}_{i,j} = \frac{\Psi_{i,j}}{\mu_{1/2}(\Psi_i)} \quad (\text{S1})$$

where  $i$  is the detector number (1–8),  $j$  is the particle number for a given experiment, and  $\mu_{1/2}(\Psi_i)$  is the median of all measurements on a single channel for the given time period. In this study, we used a time period of 5 min, as little variation in instrument conditions occurred over this time period.

After all signals have been normalized, the particle non-sphericity can be calculated by finding, for each particle, the median absolute deviation of the signal among the eight detectors by

$$\Phi_j = \mu_{1/2} \left( \left| \hat{\Psi}_{i,j} - \mu_{1/2}(\hat{\Psi}_j) \right| \right) \quad (\text{S2})$$

where  $\mu_{1/2}(\hat{\Psi}_j)$  here is the median of the normalized signal across the eight detectors for a single particle.

In an ideal case, the non-sphericity of a spherical particle is zero; that is, there is no variation in the light intensity scattered onto the eight shape detectors. Given that no instrument or particle is absolutely ideal, non-sphericity values close to zero are taken to indicate a spherical particle and values somewhat greater than zero indicate a non-spherical particle, with larger values indicating larger degrees of non-sphericity.

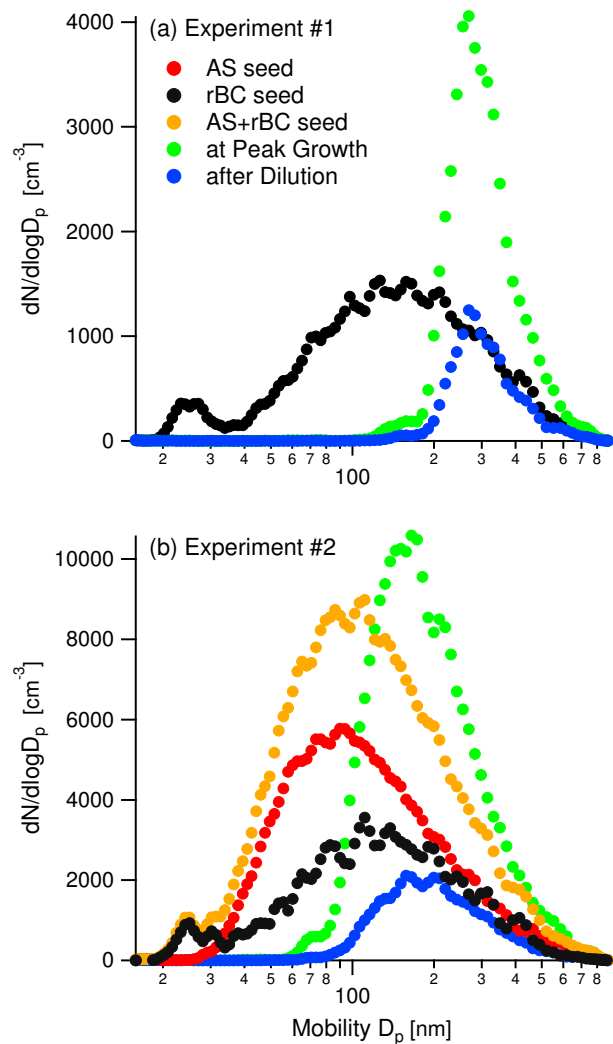


FIG. S1: Average DMA size distributions at various times throughout Experiment 1, naphthalene photooxidation in the presence of fullerene soot (rBC) seed particles, and Experiment 2, naphthalene photooxidation in the presence of rBC and ammonium sulfate (AS) seed particles. In Experiment 2 (Figure S1b), the rBC seed size distribution is determined by the difference between the AS+rBC seed and the AS seed distributions.